

# Ab-initio description of the magnetic shape anisotropy due to the Breit interaction

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A quantum-mechanical description of the magnetic shape anisotropy, that is usually ascribed to the classical magnetic dipole-dipole interaction, has been developed. This is achieved by including the Breit-interaction, that can be seen as an electronic current-current interaction in addition to the conventional Coulomb interaction, within fully relativistic band structure calculations. The major sources of the magnetic anisotropy, spin-orbit coupling and the Breit-interaction, are treated coherently this way. This seems to be especially important for layered systems for which often both sources contribute with opposite sign to the magnetic anisotropy energy. Applications to layered transition metal systems are presented to demonstrate the implications of this new approach in treating the magnetic shape anisotropy.

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Magnetic anisotropy is among the most important properties of magnetic materials in particular concerning their application in devices. When discussing the magnetic anisotropy energy of a material, denoting the difference in energy for two orientations of the magnetisation, an incoherent approach is used so far [1, 2]. On the one hand side, the dependency of the electronic structure and the associated total energy on the orientation of the magnetisation, that is induced by spin-orbit coupling (SOC), is accounted for by corresponding relativistic band structure calculations. On the other hand, the additional shape anisotropy is ascribed to the anisotropy of the magnetic dipole-dipole coupling, that is treated in a classical way. This hybrid approach is used in particular when dealing with layered transition metal systems. The pioneering and successful theoretical work on magnetic surface films by Gay and Richter [3] was followed later on by many more investigations that benefited from the extension of standard band structure schemes to account simultaneously for the presence of spin-orbit coupling and spin-polarisation, i.e. magnetisation, in a numerically reliable way. Especially interesting in this context are investigations on systems showing a competition of the SOC and shape induced contributions to the magnetic anisotropy energy. This situation is frequently encountered for magnetic multi-layer or surface layer systems for which a flip of the magnetic easy axis from out-of-plane to in-plane may be observed when the thickness of the magnetic layer is increased starting from a single mono-layer. In fact corresponding experimental findings could be reproduced by calculations based on the above mentioned hybrid scheme in the case of magnetic multi-layers [4] as well as surface layer systems [5].

In spite of the successful applications of this hybrid scheme in dealing with the magnetic anisotropy, one has to keep in mind that there is no real justification for its use. Furthermore, as there have been no coherent quantum mechanical investigations performed so far, there is no experience on the range of its applicability. It seems

that the first steps towards a coherent quantum mechanical description of the magnetic anisotropy were made by Jansen [6, 7], who pointed out that the shape anisotropy is ultimately caused by the Breit interaction – a relativistic correction to the Coulomb interaction between moving electrons [8, 9]. While Jansen performed model calculations to investigate the magnetic anisotropy energy contribution caused by the Breit interaction, first numerical investigations were done by Stiles et al. [10]. However, these authors restricted to the spin-other-orbit part of the Breit interaction that implies a current-current interaction. Performing a Gordon decomposition of the current density into a spin and orbital part [11] one gets three additional terms. Moreover, Stiles et al. investigated the pure bulk ferromagnetic metals bcc-Fe, hcp-Co and fcc-Ni for which the shape anisotropy is rather small. As a consequence, the results of this first numerical work are not very conclusive.

In this contribution we present a fully relativistic description of the magnetic anisotropy that accounts for the shape anisotropy by incorporating the full Breit interaction within the Dirac equation for magnetic solids. The scheme has been implemented by using a fully relativistic multiple-scattering or Korringa-Kohn-Rostoker (KKR) formalism. To demonstrate the power of this new approach applications to the layered systems  $\text{Fe}_n\text{Pd}_n$  and  $\text{Fe}_n/\text{Au}(001)$  are presented. The results for the magnetic anisotropy energy are compared to results obtained using the conventional hybrid approach.

The Breit-interaction [8, 9] is a correction to the Coulomb interaction between moving electrons that may be split into its magnetic and retardation part:

$$\begin{aligned} \mathcal{H}_{\text{BI}}^{(1,2)} = & -\frac{e^2}{r_{12}} \vec{\alpha}_1 \cdot \vec{\alpha}_2 \\ & + \frac{e^2}{2r_{12}} \left[ \vec{\alpha}_1 \cdot \vec{\alpha}_2 - (\vec{\alpha}_1 \cdot \vec{e}_{12}) (\vec{\alpha}_2 \cdot \vec{e}_{12}) \right]. \quad (1) \end{aligned}$$

Within this relativistic formulation, the standard Dirac matrices  $\alpha_i$  are connected to the current density operator

$j_i$  via  $\vec{j} = -|e|c\vec{\alpha}$  [11] and  $r_{12}$  denotes the distance between two electrons. The Gordon decomposition of the electronic current mentioned above is not used in the following. This implies that all coupling mechanisms connected with the Breit interaction are finally accounted for.

In the following we focus here on the magnetic part as the retardation term does not contribute on the Hartree level [12]. This allows to represent the interaction of an electron with all the other electrons by the vector potential:

$$\vec{A}(\vec{r}) = \frac{1}{c} \int d^3r' \frac{\vec{j}(\vec{r}')}{|\vec{r} - \vec{r}'|}. \quad (2)$$

Including the corresponding interaction term within electronic structure calculations done in the framework of relativistic spin density functional theory (SDFT) [13] leads to the following single particle Dirac Hamiltonian:

$$\mathcal{H}_D = -ic\vec{\alpha} \cdot \vec{\nabla} + \beta mc^2 + V_{\text{eff}}(\vec{r}) + \beta \vec{\Sigma} \cdot \vec{B}(\vec{r}) + e\vec{\alpha} \cdot \vec{A}(\vec{r}), \quad (3)$$

with the corresponding total energy  $E$  given by:

$$E = T_s + E_{\text{ext}} + E_H^C + E_H^T + E_{\text{xc}} \quad (4)$$

Here the first three and the last terms have their usual meaning, i.e. they stand for the kinetic energy ( $T_s$ ), the electron-nucleus ( $E_{\text{ext}}$ ) and electron-electron ( $E_H^C$ ) Coulomb interaction and the exchange-correlation ( $E_{\text{xc}}$ ) contributions. The fourth term:

$$E_H^T = -\frac{e^2}{2c} \int d^3r \int d^3r' \frac{\vec{j}(\vec{r}) \cdot \vec{j}(\vec{r}')}{|\vec{r} - \vec{r}'|}, \quad (5)$$

going beyond standard SDFT, is a Hartree-like term due to the Breit or current-current interaction. As mentioned above the retardation term in Eq. (1) does not contribute to  $E_H^T$  [12]. This could indeed be verified by our numerical results.

As usually done for total energy calculations, the kinetic energy term in Eq. (4) can be eliminated by making use of the Hamiltonian in Eq. (3).

When calculating the vector potential  $\vec{A}(\vec{r})$  it is advantageous to decompose the integration regime in Eq. (2) into the central atomic cell  $i$  and its surrounding giving rise to the on- and off-site contribution, respectively, for the vector potential within cell  $i$ :

$$\vec{A}_i(\vec{r}) = \vec{A}_i^{\text{on}}(\vec{r}) + \vec{A}_i^{\text{off}}(\vec{r}). \quad (6)$$

The on-site contribution  $\vec{A}_i^{\text{on}}$  can be determined directly from the currents within atomic cell  $i$ , on the basis of Eq. (1). The off-site contribution  $\vec{A}_i^{\text{off}}$  from all other sites can be obtained by applying the common far field approximation

$$\vec{A}_i^{\text{off}}(\vec{r}) = \sum_{j \neq i} \frac{\vec{M}_j \times (\vec{r} - \vec{R}_j)}{|\vec{r} - \vec{R}_j|^3}, \quad (7)$$

where the total magnetic moment  $\vec{M}_j$  represents the current distribution in atomic cell  $j$ . The lattice summation in Eq. (7) is dealt with by an Ewald summation technique in the case of two- or three-dimensional periodic systems.

Using a band structure method based on a decomposition of the system into atomic cells, as for the KKR-method used here, Eq. (3) has to be solved in a first step for isolated atomic cells (single-site problem). For this purpose it is helpful to expand the vector potential by means of spherical harmonics. For the Breit-interaction term, the last term in Eq. (3), one obtains:

$$\mathcal{H}_{\text{BI}} = e \sum_{m_\alpha} \alpha_{m_\alpha} \sum_{\ell_A m_A} A_{\ell_A m_A}^{-m_\alpha}(r) Y_{\ell_A}^{m_A}(\hat{r}). \quad (8)$$

In the implementation presented here the atomic sphere approximation (ASA) together with a restriction to collinear magnetism has been applied, i.e. within an atomic cell one has  $V_{\text{eff}}(\vec{r}) = V(r)$  and  $\vec{B}(\vec{r}) = B(r)\hat{e}_M$ , with the orientation of the magnetic moments  $\hat{e}_M$  that fixes the local  $z$ -axis. In line with these geometrical simplifications  $\vec{A}(\vec{r})$  is restricted to have rotational symmetry around  $\hat{e}_M$  with  $\vec{A}(\vec{r})$  pointing everywhere in tangential direction, i.e.  $\vec{A}(\vec{r}) = A(r, \theta)\hat{e}_\phi$  implying  $A_{\ell_A-1}^{+1}(r) = -A_{\ell_A+1}^{-1}(r)$  and all other terms being zero. (Further technical details will be given elsewhere [14]).

With the single-site Dirac equation being solved, the electronic structure of the investigated system can be calculated by means of the standard relativistic multiple scattering or KKR-method [15]. Calculating the total energy  $E(\hat{n})$  on the basis of Eq. (4) for two different orientations of the magnetisation  $\hat{n}$  and  $\hat{n}'$ , respectively, gives the corresponding magnetic anisotropy energy  $\Delta E$  as the difference  $E(\hat{n}) - E(\hat{n}')$ .

As an application of the approach sketched above we consider the geometrically simple case of a free-standing bcc Fe monolayer with a lattice constant of bulk Fe ( $a = 2.87 \text{ \AA}$ ). The magnetic moment  $\vec{M}$  was taken to point out-of-plane, i.e. along the  $z$ -axis of the system. The top left panel of Fig. 1 shows the resulting radial vector functions  $A_{\ell_A m_A}^{-m_A}(r)$  for  $m_A = 1$  with the on- and off-site parts to  $\vec{A}(\vec{r})$  indicated by black and red lines, respectively,

The top right panel of Fig. 1 shows the resulting radial dependence of the  $\phi$ -component  $A_\phi^{\text{on}}$  along the  $x$ -direction (black line) together with  $A_\phi^{\text{off}}$  obtained via Eq. (7) (red line). Applying the far field approximation of Eq. (7) also to the on-site term results in the blue curve which is divergent at the origin. In the outermost region of an atomic sphere, however, the far field approximation to  $\vec{A}(\vec{r})$  is already very good justifying the use of Eq. (7) also for nearest neighbouring atoms.

In order to verify the correctness of  $A_\phi^{\text{on}}$  when calculated directly via  $\mathcal{H}_{\text{BI}}$  (see Eq. (1)) we also determined the current density  $\vec{j}(\vec{r})$  within an Fe sphere and then

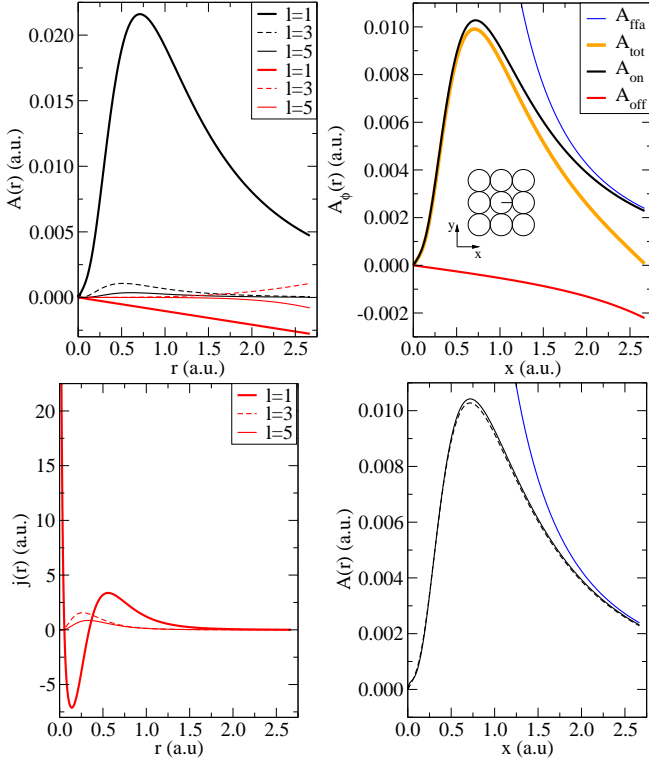


FIG. 1: Vector potential and current density for Fe in a free-standing monolayer. Top left: On- (black) and off-site (red) contributions to the vector potential functions  $A_{\ell_A m_A}^{-m_A}(r)$  for  $\ell_A = 1, 3, 5$  and  $m_A = 1$  in atomic units. Top right: On- (black) and off-site (red)  $A_\phi$  component along the  $x$ -direction ( $\theta = \frac{\pi}{2}, \phi = 0$ ).  $\vec{A}^{\text{on}}$  is compared to the far field approximation  $\vec{A}^{\text{ffa}}$  (blue curve). Bottom left: radial current density functions  $j_{\ell m}^{-m}(r)$  for  $\ell = 1, 3, 5$  and  $m = 1$ . Bottom right: comparison of  $A_\phi$  along  $x$  calculated via  $\mathcal{H}_{\text{BI}}$  (full line) and from the current density (dashed line).

computed  $A_\phi^{\text{on}}$  via Eq. (2). The bottom left panel of Fig. 1 shows the radial electronic current density distribution functions  $j_{\ell m}^{-m}(r)$  (defined in analogy to  $A_{\ell_A m_A}^{-m_A}$  in Eq. (8)) for an Fe atom and the bottom right panel of Fig. 1 presents a comparison of  $A_\phi^{\text{on}}$  resulting from  $j_{\ell m}^{-m}(r)$  (dashed line) and obtained from  $\mathcal{H}_{\text{BI}}$ . As one can see both approaches give nearly identical results.

A further impression of the spatial variation of  $\vec{A}(\vec{r})$  and  $\vec{j}(\vec{r})$  is given in Fig. 2 showing the vector fields in the  $xy$ -plane and their color-coded amplitude in the  $xz$ -plane. The figure reflects the rotational symmetry of  $\vec{A}(\vec{r})$  imposed by the use of the ASA (atomic sphere approximation) and the alignment of the magnetisation along the  $z$ -direction.

For the anisotropy energy  $\Delta E = E(\hat{x}) - E(\hat{z})$  of the free-standing bcc Fe monolayer we obtain  $-0.063$  meV with the sign indicating that the magnetisation favours an in-plane orientation.  $\Delta E$  can be decomposed into  $\Delta E_{\text{SOC}} = 0.096$  meV preferring an out-of-plane magnetic easy axis and a dominating  $\Delta E_{\text{BI}} = -0.159$  meV

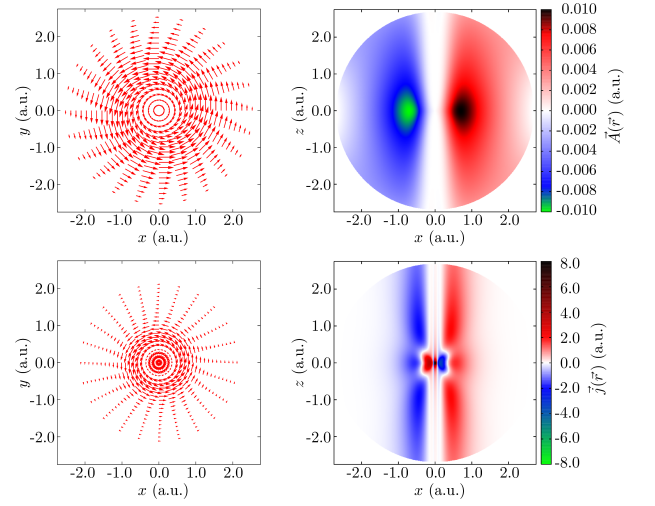


FIG. 2: Top: Vector potential  $\vec{A}(\vec{r})$  of Fe within the  $xy$ -plane with  $z = 0$  (left) and within the  $xz$ -plane with  $y = 0$  (right) for the magnetisation along the  $z$ -axis. Bottom: Corresponding current density  $\vec{j}(\vec{r})$  within the  $xy$ -plane (left) and within the  $xz$ -plane (right).

causing the magnetisation to lie in-plane. Here,  $\Delta E_{\text{BI}} = \Delta E - \Delta E_{\text{SOC}}$  has been obtained by performing calculations with (SOC+BI) and without (SOC only)  $\mathcal{H}_{\text{BI}}$  giving  $\Delta E$  and  $\Delta E_{\text{SOC}}$ , respectively. For the classical dipolar shape anisotropy  $\Delta E_{\text{dd}}$  we get  $-0.169$  meV which agrees astonishingly well with the quantum mechanical  $\Delta E_{\text{BI}}$ .

A second application dealt with the multilayer systems  $\text{Fe}_n\text{Pd}_n$ . For these a fcc structure with (001)-oriented atomic layers has been assumed, i.e. with the  $z$ -axis perpendicular to the Fe- and Pd-layers, respectively. For the special case  $n = 1$  this corresponds to the CuAu-structure.

The impact of the Breit-interaction on the electronic structure leads to a competition of the various contributions to the magnetic anisotropy energy  $\Delta E$  of  $\text{Fe}_n\text{Pd}_n$  when the parameter  $n$  is varied. Using the conventional approach that accounts only for  $\Delta E_{\text{SOC}}$  within the electronic structure calculations leads to a strong contribution that favors an out-of-plane orientation of the magnetisation for  $n = 1 - 6$  (see Fig. 3). The additional classical shape anisotropy contribution  $\Delta E_{\text{dd}}$  gives rise to a contribution that favours an in-plane orientation of the magnetisation. As one may expect,  $\Delta E_{\text{dd}}$  of  $\text{Fe}_n\text{Pd}_n$  is primarily determined by the magnetic moments within the Fe layers that amounts to be between  $2.74$  and  $2.95 \mu_B$  for all values of  $n$  considered here. The induced moments on the Pd layers that are in the range  $0.005$  to  $0.330 \mu_B$  are only of minor importance. As a result of this  $\Delta E_{\text{dd}}$  increases nearly linearly with  $n$  for the range of  $n$  considered here. For  $n = 5$  and presumably also for  $n > 6$  the magnetic dipole-dipole term exceeds the SOC-induced term leading to a flip of the magnetic

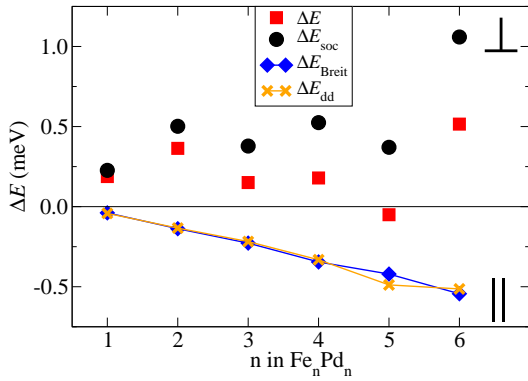


FIG. 3: Magnetic anisotropy energies for repeating  $\text{Fe}_n\text{Pd}_n$  multilayers as function of  $n$ : total magnetic anisotropy energy  $\Delta E$  (squares) and its decomposition into magneto-crystalline part  $\Delta E_{\text{SOC}}$  (circles) and Breit part  $\Delta E_{\text{BI}}$  (diamonds) which is compared to its classical approximation (crosses).

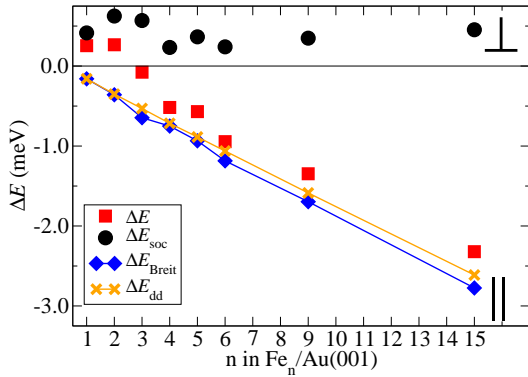


FIG. 4: As for Fig. 3, but for the surface layer system  $\text{Fe}_n/\text{Au}(001)$ .

easy axis from out-of-plane to in-plane.

Calculating the anisotropy energy  $\Delta E$  on the basis of the coherent SOC+BI scheme,  $\Delta E$  follows qualitatively the variation of the SOC-induced magnetic anisotropy energy with  $n$  and implies also a flip of the magnetic easy axis (see Fig. 3). To allow for a direct comparison of the two approaches the difference  $\Delta E_{\text{BI}}$  of SOC+BI and the SOC-only scheme is shown as well in Fig. 3. As can be seen,  $\Delta E_{\text{BI}}$  is very close to the classical  $\Delta E_{\text{dd}}$ . This result obviously justifies the use of the conventional classical approach for the shape anisotropy used so far. In particular the conventional scheme seems to reproduce the quantum mechanical result not only in a qualitative but in general also quantitatively in a satisfying way. Obviously, only for rather short interatomic distances one has to be aware of possibly pronounced deviations between the classical and quantum mechanical approaches.

These conclusions are confirmed by results obtained for the closely related multilayer systems  $\text{Fe}_n\text{Pt}_n$ ,  $\text{Co}_n\text{Pd}_n$  and  $\text{Co}_n\text{Pt}_n$  as well as first applications to surface layer systems. Fig. 4 shows corresponding results for the sys-

tem  $\text{Fe}_n/\text{Au}(001)$  that exhibits a flip of the easy axis from out-of-plane to in-plane for around three monolayers of Fe (see also Ref. [5]). Again the contribution  $\Delta E_{\text{BI}}$  to the total magnetic anisotropy energy  $\Delta E$  is found to be very close to the classical result  $\Delta E_{\text{dd}}$ . Only for rather large thicknesses a noteworthy deviation of the two can be seen in Fig. 4.

In summary the Breit interaction has been incorporated within fully relativistic band structure calculations for magnetic layered systems. This development gives access to an ab-initio calculation of the magnetic shape anisotropy energy using a coherent approach that accounts simultaneously for spin-orbit coupling and the Breit interaction. First applications of this new approach to the systems  $\text{Fe}_n\text{Pd}_n$  and  $\text{Fe}_n/\text{Au}(001)$  were presented. Taking the difference of the calculated magnetic anisotropy energy obtained via the combined SOC+BI - and the SOC-only approaches, the contribution due to the Breit interaction could be separated. For all systems investigated so far it was found that the resulting Breit contribution is very close to the classical result calculated on the basis of the magnetic dipole-dipole interaction. This result can be explained to some extent by the fact that it is the magnetic part of the Breit interaction that gives rise to the shape anisotropy while the retardation term does not contribute on the Hartree-level.

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